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Synthesis and Binding Constants for Poly-Amidoxime Uranyl Complexes for Sequestering Uranium from Seawater

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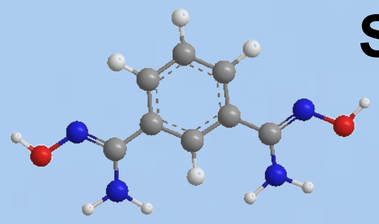

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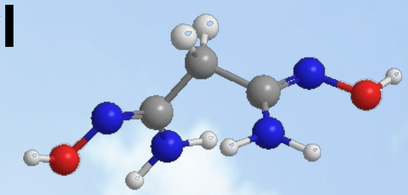

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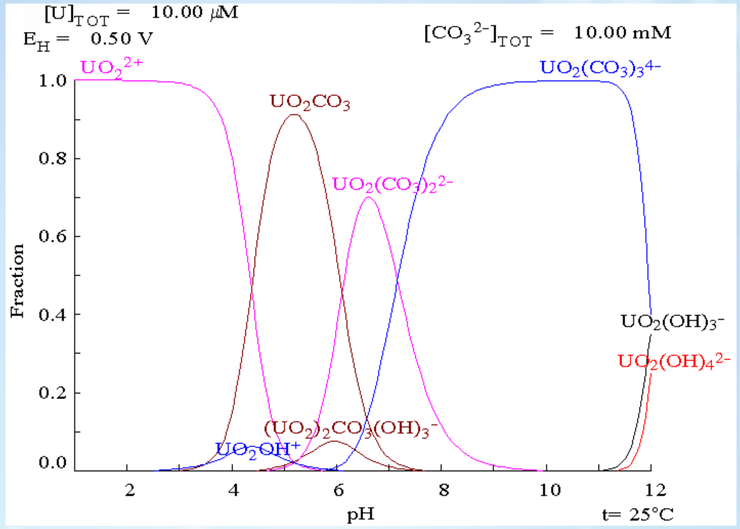
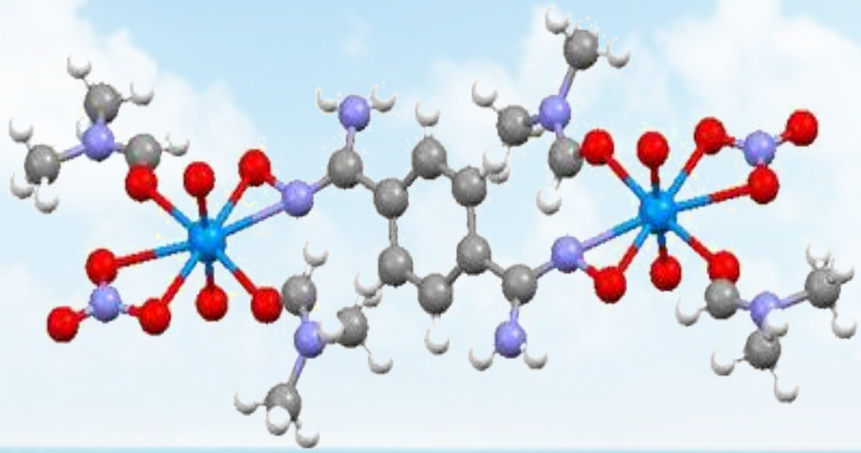
How do we Sequester Uranium?

- There is estimated to be upwards of 1000 times as much uranium in seawater then available terrestrially
- If harnessed, could provide energy for the next 100 years at our current nuclear growth.
- With Multi-Topic amidoximes, we could potentially increase effectiveness of currently used molecules
- Due to uranium's low concentration, estimated at 3.3 ppb, our ligands must be able to selectively bind to uranium and not competitor ions
- Due to the high stability of uranyl-carbonate species, these ligands must be highly competitive
- Must be able to withstand high pH conditions to be reusable in sequestering process

Results

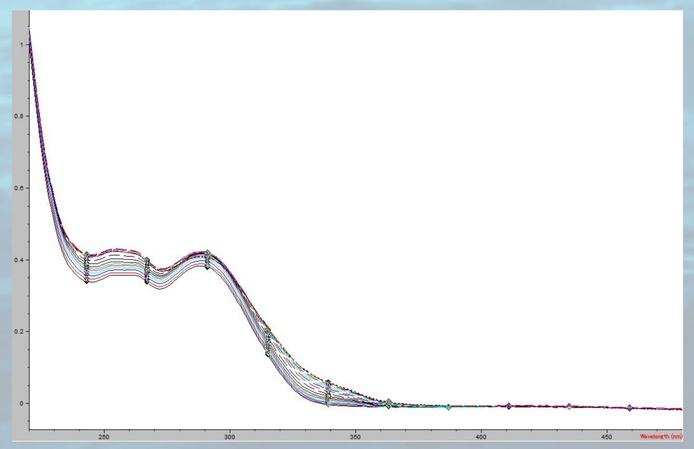
- Protonation constants have been determined for several of our ligands, while the binding constants are being processed.
- In solution, our ligands have shown a wide array of speciation possibilities, each having its own unique properties.
- Crystal structures for two species have been obtained, one showing a rather surprising configuration.

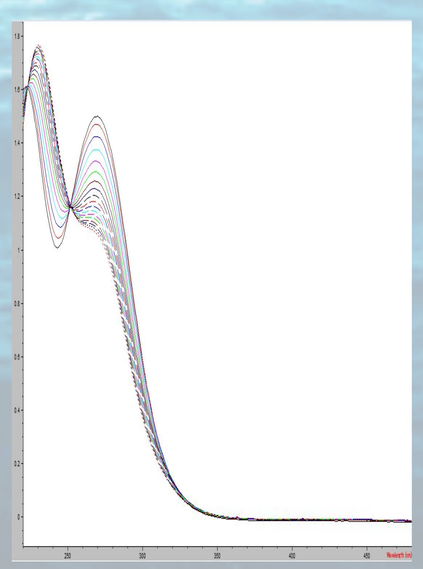


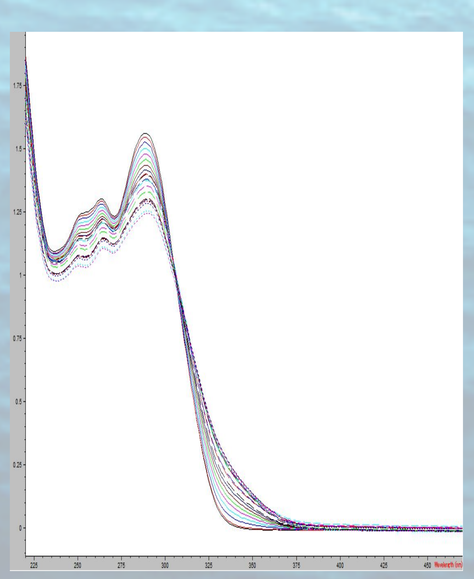


Materials and Methods

- All amidoximes were prepared by reacting their corresponding nitriles with 2.3 molar eq. free hydroxylamine.
- Purified through HPLC, recrystallization, or column chromatography.
- pKa's of the ligands we determined through NMR, UV/Vis, and potentiometric titration methods and fit using the non-linear regression program HypQuad, HypSpec, and HypNMR.
- Initial solutions contained .1mM ligand, .5M Ionic Strength, and was either titrated with .1M NaOH or .01M HCl
- Uranyl Nitrate Hexahydrate was used as our uranyl cation source.
- Binding constants were determined through UV/Vis and Potentiometric titrations.
- Initial solutions contained .1mM ligand, .044mM Uranyl, and were titrated with .1M NaOH







Conclusions and Implications

- Our ligands have shown to have a high binding affinity to uranium in solution, being able to form larger structures based around the uranium
- Binding constants are being determined for many of the ligands, with experiments done on three of our potential molecules.
- We look to try and design a molecule that will hopefully be able to encapsulate a uranium ion in solution, reulting in a potentially very stable 1:1 binding ratio.
- The ongoing hunt for crystals showing how a single molecule can bind and influence the resulting metal-to-ligand ratio is ongoing, with well over one hundred crystal attempts being set up.
- More complex solutions will be analyzed in an attempt to determine the binding constants of more complex structures.

Literature Cited

Dalton Trans., 2014, 43, 551

Dalton Trans., 2012, 41, 11579

Acknowledgments

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